LLNL-CONF-698945

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July 29, 2016

15th International Conference on X-Ray Lasers
Nara, Japan
May 22, 2016 through May 27, 2016

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Using the X-FEL to drive gain in K-shell and L-shell systems using photo-ionization and photo-excitation of inner-shell transitions

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Abstract. Five years ago an inner-shell X-ray laser was demonstrated at 849 eV (1.46 nm) in singly ionized neon gas using the X-FEL at 960 eV to photo-ionize the 1s electron in neutral neon followed by lasing on the 2p – 1s transition in singly-ionized neon. This work was done at the SLAC Linac Coherent Light Source (LCLS) by a multi-laboratory team led by Nina Rohringer and published in the January 26, 2012 issue of Nature. It took decades to demonstrate this scheme because it required a very strong X-ray source that could photo-ionize the 1s (K shell) electrons in neon on a time scale comparable to the intrinsic auger lifetime in the neon, which is typically 2 fsec. In this work we model a neon inner shell X-ray laser driven by the XFEL at LCLS and show how we can improve the efficiency of the neon laser and reduce the drive requirements by reducing the pulse duration to 1-ps and tuning the XFEL to the 1s-3p transition in neutral neon in order to create gain on the 2p-1s line in neutral. We also show how the XFEL could be used photo-ionize L-shell electrons to drive gain on n=3-2 transitions in singly-ionized Ar and Cu plasmas. These bright, coherent, and monochromatic X-ray lasers may prove very useful for doing high-resolution spectroscopy and for studying non-linear process in the X-ray regime.

1 Introduction

Scientists have proposed schemes to achieve lasing at shorter wavelengths since the invention of the laser. In the 1960's, Duguay and Rentzepis, proposed using photo-ionization to create an X-ray laser on the inner shell K-α line in sodium vapour [1]. In the 1970's Ray Elton [2] discussed the challenges of making quasi steady state inner-shell K-α lasers in Si, Ca, and Cu. The dream of demonstrating an inner-shell X-ray laser was realized at the SLAC Linac Coherent Light Source (LCLS) in 2011 when the X-ray free electron laser (XFEL) at 960 eV was used to photo-ionize the K-shell of neutral neon gas and create lasing at 849 eV in singly ionized neon gas [3].

An alternative approach for creating X-ray lasers was the idea of a resonantly photo-pumped laser where a strong emission line in one material could be used to photo-excite a transition in another material and create lasing. A classic example is the Na-pumped Ne X-ray laser scheme proposed

40 years ago by Vinogradov and colleagues [4-5] that used the strong Na He-α line at 1127 eV to resonantly photo-pump the Ne He-γ line and lase on the 4f – 3d transition at 23.1 nm in He-like Ne. This scheme was studied extensively and weak gain [6] was inferred in several experiments. The difficulty with this type of scheme was creating a sufficiently strong pump line. With the availability of strong XFEL sources the pump line in the traditional photo-pumped schemes can be replaced with an XFEL that is tuned to the appropriate resonance. Since the resonant photo-pumped scheme selectively pumps a transition it offers the potential for higher gain and lower drive intensity than the photo-ionization pumping.

In this paper we look at the advantages and challenges of using the XFEL to resonantly photo-pump the 1s-3p line in neutral neon as a mechanism for creating gain on the K-α line in Ne and compare this with the photo-ionization pumping that has already been demonstrated. We show that with the use of a sufficiently short XFEL pulse (1-fsec) the resonant photo-excitation could reduce the XFEL flux requirements by several orders of magnitude. We then look at how the inner-shell X-ray laser can be extended to lasing on L-shell transitions in Ar and Cu. For Ar we consider an XFEL pulse that photo-ionizes the 2p or 2s electrons and creates lasing on the 3s – 2p or 3p – 2s transitions. In the case of Cu we consider an XFEL pulse that photo-ionizes the 2p electron and creates lasing on the strong 3d – 2p transitions near 1 keV.

2 Modelling the inner-shell Ne laser

In the LCLS experiments a strong XFEL beam was tuned above the K-edge of neutral Ne I to photo-ionize the 1s electron as shown in Fig. 1. An excited state of singly ionized Ne II is created that has a missing 1s electron denoted by (1s) from the closed K and L shell denoted by [KL]. This excited state lases to the ground state of Ne II by emitting an X-ray on the 2p – 1s transition at 848.6 eV. The experiment starts with cold, neutral neon gas that is in the Ne I ground state. The lower laser state is initially unoccupied. Even though the natural lifetime of the laser transition is 135 fsec, the challenge with this scheme is that the Auger lifetime of the upper laser state is 2.3 fsec. This scheme requires a very short pulse duration in the fsec regime for photo-ionizing the 1s electron.

Figure 1 shows the resonant photo-pumping mechanism for driving the inner-shell neon laser. The XFEL is tuned to the 1s – 3p transition in Ne I at 867.63 eV creating a large population in the [KL] (1s) 3p level. This level can

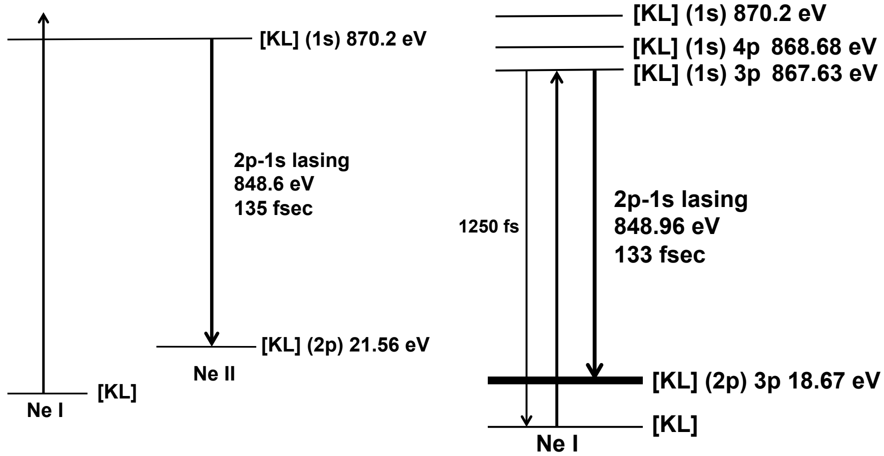


Fig. 1. Energy level diagrams for the photo-ionization (left) and photo-excitation (right) driven inner-shell neon X-ray laser.

then lase to the lower [KL] (2p) 3p level state by emitting X-rays on the 2p – 1s transition centered at 848.96 eV. The lower level state is split into multiple levels so there are five X-ray lines emitted that are spread over a range of 848.67 - 849.25 eV. The total gain is calculated by summing the gain of the 5 lines. The upper laser level has an Auger lifetime of 2.3 fsec which implies a line-width of 0.3 eV on the lasing transition. The main difference between this scheme and the photo-ionization scheme is that lasing occurs in neutral Ne I. The lasing energies differ by about 0.4 eV so it is possible to observe the difference in lasing energy between the two pumping mechanisms. The potential advantage of this scheme is that the photo-excitation cross-section is about 18 Mbarns compared to 0.3 Mbarns for the photo-ionization scheme. The question we want to address in this paper is how we can take advantage of this much larger excitation rate to reduce the drive requirements on the XFEL source.

To model the photo-ionization and photo-excitation schemes we created a simple atomic model of the levels shown in Fig. 1. We used the kinetics code Cretin [7] to model the kinetics and gain of the system under various conditions. For the baseline XFEL beam we assume the XFEL beam has 10^{12} photons in a 0.9 eV line-width focused to a 1-μm diameter. In our previous work we had studied the nominal LCLS conditions that used a 100-fsec full-width half-maximum (FWHM) Gaussian pulse [8]. In this work we examine using a shorter 1-fsec FWHM pulse which produces much higher gain than the 100-fs pulse used in the LCLS experiments [3,8]. The XFEL was designed to have a bandwidth of 0.1%, as we are assuming, even though the current bandwidth is larger by a factor of 5-10. The bandwidth has minimal impact on the photo-ionization mechanism but the strength of the photo-excitation mechanism is inversely proportional to the bandwidth. One challenge with the photo-excitation scheme is understanding the validity of the kinetics model and how to include the photo-excitation rate in the line-width calculation of the gain. Currently the stimulated rate is included in the kinetics but not in the line-width which means there are no Stark sidebands or broadening. The XFEL energy is set at 875 eV for modelling the photo-ionization scheme and 867.6 eV for modelling the photo-excitation scheme.

Figure 2 shows the predicted gain versus time for both schemes. To understand the sensitivity to the XFEL flux a series of calculations were done using a multiplier between 1.0 (nominal) and 0.001 on the nominal XFEL flux described above. For the nominal XFEL flux the peak gains are 910 cm^{-1} at -0.5 fsec for the photo-ionization and 703 cm^{-1} at -1.1 fsec for photo-excitation. By comparison the peak gain were predicted to be 42 and 62 cm^{-1}, respectively, using a 100-fs XFEL pulse with the same number of photons. As the XFEL intensity is reduced the gain for the photo-ionization drops quickly but one notices that the peak gain for the photo-excitation scheme drops from 703 cm^{-1} to 639 cm^{-1} as the XFEL drive flux is reduced by a factor of 100. Also the peak of the gain moves to a time of -0.2 fsec, indicating near optimum drive conditions since the peak of the XFEL pulse is defined as time = 0. The estimated Rabi width associated with the resonant pumping is 1 eV for this case which is similar to the linewidth of the XFEL line. As the XFEL flux drops further the gain drops more quickly and occurs after the peak of the XFEL flux indicating the flux is below ideal drive conditions. This figure

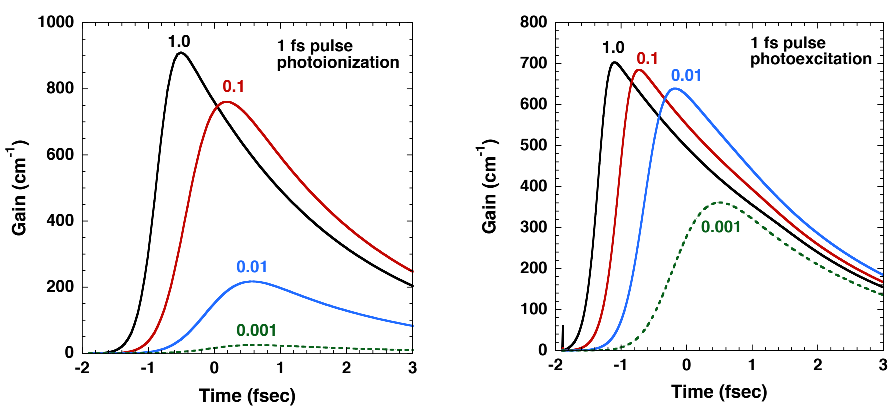


Fig. 2. Gain versus time for the neon X-ray laser driven by a 1-fs duration XFEL comparing the photo-ionization and photo-excitation mechanisms. The XFEL intensity is varied by using a multiplier between 1.0 (nominal) and 0.001. The peak of the XFEL pulse is defined as time = 0.

shows that the photo-excitation mechanism offers the potential to reduce the XFEL drive by one to two orders of magnitude as compared with the photo-ionization mechanism. This advantage could enable smaller facilities to drive inner shell X-ray lasers or allow facilities such as LCLS to drive even higher energy X-ray lasers with the current XFEL fluxes.

3 Modelling Ar and Cu L-shell X-ray lasers

Given the success of the K-shell neon X-ray laser it should be possible to demonstrate inner-shell X-ray lasers in other principal shells such as the L and M shells. One promising candidate to consider is neutral argon gas. Figure 3 shows the energy level diagram for using an XFEL above the L-shell edge of neutral Ar I to create a L-shell hole in singly ionized Ar II. If an XFEL was tuned between the two L-edges at 250 and 326 eV one could create a 2p hole that would result in lasing on the 3s-2p transitions at 219 and 221 eV. If the XFEL drive was tuned above the L-edge at 326.3 eV then one would have holes in both the 2s and 2p shells that would result in lasing on the 3p-2s transitions at 310.4 and 310.6 eV as well as the 3s-2p transition. It would be very interesting to tune the XFEL from low to high energy and watch the 3s-2p lasing turn on followed by lasing on both lines. Fig. 3 shows the gain versus time for the Argon 3s-2p X-ray laser line at 219 eV driven by a 260 eV XFEL with 10^{12} photons in a 1-fsec pulse focused to 1-μm diameter spot. The 219 eV line has twice the gain of the 221 eV line. One observes that the peak

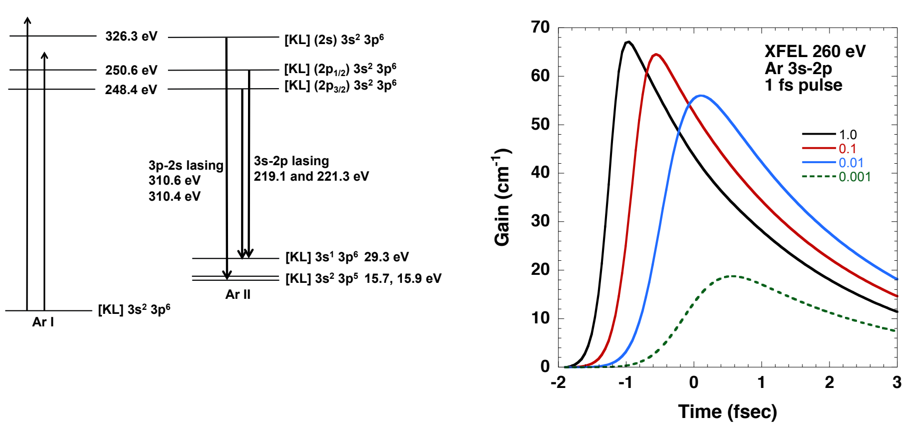
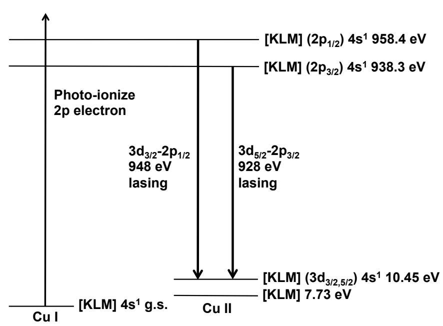


Fig. 3. Energy level diagram (left) for the photo-ionization driven inner-shell argon X-ray laser. Gain versus time (right) for the 3s-2p line at 219 eV in the argon X-ray laser driven by the photo-ionization mechanism using a 1-fs duration 260 eV XFEL drive pulse. The XFEL intensity is varied by using a multiplier between 1.0 (nominal) and 0.001. The peak of the XFEL pulse is defined as time = 0.

gain of 67 cm^{-1} falls very slowly to 56 cm^{-1} as the XFEL flux is reduced by a factor of 100. By comparison, using a 330 eV XFEL pulse the 3p-2s line at 310 eV has a peak gain of about 30 cm^{-1}.

Figure 4 shows the energy level diagram for using an 1 keV XFEL whose energy is above the L-shell edge of neutral Cu I to create a L-shell hole in singly ionized Cu II and create lasing on the strong 3d-2p lines at 928 and 948 eV. The gain at 928 eV is predicted to be about twice the gain at 948 eV so Fig. 4 shows the gain versus time for the Copper 3d-2p X-ray laser line at 928 eV driven by a 1000 eV XFEL with 10^{12} photons in a 10-fsec pulse focused to 1-µm diameter spot. The gain peaks at 136 cm^{-1} and falls slowly as the XFEL intensity is reduced. As an alternative, photo-excitation of the 2p-4d transition in Cu I would also create lasing on the 3d-2p line in Cu I and might require an even lower XFEL intensity.



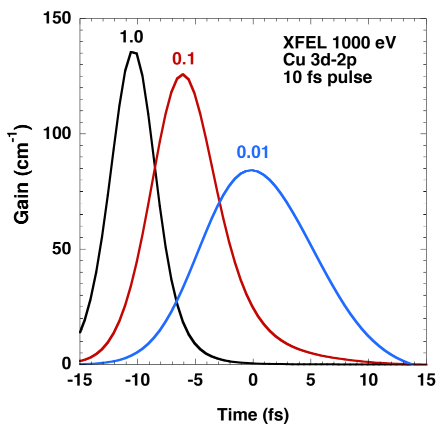


Fig. 4. Energy level diagram (left) for the photo-ionization driven inner-shell copper X-ray laser. Gain versus time (right) for the 3d-2p line at 928 eV in the copper X-ray laser driven by the photo-ionization mechanism using a 10-fs duration 1000eV XFEL drive pulse. The XFEL intensity is varied by using a multiplier between 1.0 (nominal) and 0.01. The peak of the XFEL pulse is defined as time = 0.

4 Conclusions

In this paper we model the neon inner shell X-ray laser under similar conditions to those used at LCLS. We show how we can improve the efficiency of the neon laser and reduce the drive requirements by tuning the XFEL to the 1s-3p transition in neutral neon in order to create gain on the 2p-1s line in neutral neon. We present the sensitivity to the drive intensity, pulse duration, and line-width of the XFEL to better understand how to optimize this inner shell laser by understanding the trade-offs between using photo-ionization versus photo-excitation to drive gain in these systems. We show that with the use of a sufficiently short XFEL pulse (1-fsec) the resonant photo-excitation could reduce the XFEL flux requirements by several orders of magnitude. We also discuss how photo-ionization of L-shell electrons can be used to create lasing on n=3-2 transitions in materials such as Ar at 219, 221, and 310 eV and Cu at 928 and 948 eV.

Acknowledgements. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. M. A. Duguay and P. M. Rentzepis, Appl. Phys. Lett. **10**, 350–352 (1967).
2. R. C. Elton, Appl. Opt. **14**, 2243- 2249 (1975).
3. N. Rohringer, D. Ryan, R. A. London, M. Purvis, F. Albert, J. Dunn, J. D. Bozek, C. Bostedt, A. Graf, R. Hill, S. P. Hau-Riege, and J. J. Rocca, Nature 481, 488–491 (2012).
4. A. V. Vinogradov, I. I. Sobelman, and E. A. Yukov, Sov. J. Quantum Electron. **5**, 59–63 (1975).
5. J. Nilsen, J. H. Scofield, and E. A. Chandler, Appl. Opt. **31**, 4950 – 4956 (1992).
6. J. Nilsen and E. Chandler, Phys. Rev. **A 44**, 4591-4598 (1991).
7. H. A. Scott, JQSRT **71**, 689–701 (2001).
8. J. Nilsen, Matter and Radiation at Extremes **1**, 76-81 (2016).